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Numerical Simulation of Spouted Bed Reactors using Process Engineering Models: Application to Coal Gasification

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NUMERICAL SIMULATION OF SPOUTED BED REACTORS USING PROCESS ENGINEERING MODELS: APPLICATION TO COAL GASIFICATION

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ABSTRACT

A spouted bed reactor operating at high temperature has been modelled using one dimensional models based on process engineering concepts. The process of coal gasification has been selected to demonstrate the models' achievements and predictions have been compared to previous spouted bed reactor experimental results.

INTRODUCTION

Spouted bed reactors are an alternative to conventional gas-solid contactors such as fluidised bed reactors for coarse particles. They are used in various industrial operations such as drying, granulation, coating, heterogeneous catalysis, gasification of biomass and coal, etc. The successful design and operation of a gas-solid spouted bed reactor depend on the ability to predict system hydrodynamics, as well as coupled heat and mass transfer with chemical reaction rates. Since direct experimental measurement of velocities, temperatures and species concentrations inside the bed is technically difficult, numerical simulation turns out to be a powerful tool to investigate the reactor behaviour and performance.

The most commonly used approaches for modeling gas–solid systems are the discrete element method, the two–fluid method and the process engineering methods. The two former approaches have been mostly developed over the past 10 years and have demonstrated their superiority for hydrodynamic simulation (Krzywanski et al., 1992; Huilin et al., 2001; Kawaguchi et al., 2000). However, they require large computational resources and long simulation times and are generally used for modeling non-reacting or simple chemical systems (Limtrakul et al., 2004), since coupling heat and mass transfer is a difficult task in these approaches. Conversely, models based on a process engineering approach can handle complex chemical reaction systems; however, most of the models proposed to date are only one-dimensional models that often neglect some important heat transfer phenomena such as radiation and heat transfer at walls.

This study aims at modelling hydrodynamics, heat and mass transfer (with reaction kinetics) in a spouted bed reactor through a process engineering approach. The process of coal gasification has been selected to demonstrate the prediction capability of our models, and predictions have been compared to previous modelling

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results. The experimental results selected in this work are from Lucas et al. (1998), who developed a non isothermal model of a spouted bed gasifier, and from Salam and Bhattacharya (2006), who studied charcoal gasification in two different configurations of spouted bed, comparing bed temperatures and species concentrations at the reactor exit, as well as gasification efficiencies.

MODEL FORMULATION

Mathematical Formulation

Assuming plug flow of both upward gas and upward and downward solids in the two regions of the bed, namely the spout and the annulus, continuity and mass balances were written over a differential height of reactor (*Table 1*). Four energy balances were also considered, which correspond to gas and particle temperature fields in each hydrodynamic region of the bed. Heat transfer modes considered were convection, gas-particle heat exchange, conduction (bed effective conductivity), radiation and wall heat transfer.

Table 1: Mathematical formulation

Mass balance for the gaseous component j	
Spout region	$A_s U_s \frac{dC_{sj}}{dz} = r_{sj} A_s$
Annulus region	$A_a U_a \frac{dC_{aj}}{dz} = r_{aj} A_a + \left[A_a \frac{dU_a}{dz} + U_a \frac{dA_a}{dz} \right] \left(C_{sj} - C_{aj} \right)$
Energy balance	
Spout region	
Gas phase	$\rho_g C_{pg} U_s \frac{dT_{sg}}{dz} = h_{pg} \frac{6(1-\varepsilon_s)}{d_p} \left[T_{sp} - T_{sg} \right] + \sum \left(-\Delta H_r \right) \Re_g$
Solid phase	$\rho_p C_{pp} V_s A_s \frac{dT_{sp}}{dz} = A_s h_{pg} \frac{6(1-\varepsilon_s)}{d_p} \left[T_{sg} - T_{sp} \right] + \rho_p C_{pp} \left[A_s \frac{dV_s}{dz} + V_s \frac{dA_s}{dz} \right] \left(T_{sg} - T_{sp} \right) + \sum \left(-\Delta H_r \right) \Re_p$
Annulus region	
Gas phase	$\rho_g C_{pg} U_a A_a \frac{dT_{ag}}{dz} = A_a h_{pga} \frac{6(1-\varepsilon_a)}{d_p} \left[T_{ap} - T_{ag} \right] + \rho_g Cp_g \left[A_a \frac{dU_a}{dz} + U_a \frac{dA_a}{dz} \right] \left(T_{ag} - T_{ag} \right) + \sum \left(-\Delta H_r \right) \Re_g$
Solid phase	$k_{ea} A_a \frac{d^2 T_{ap}}{dz^2} = A_a h_{pga} \frac{6\left(1-\varepsilon_a\right)}{d_p} \left[T_{ap} - T_{ag} \right] + \rho_p Cp_p V_a A_a \frac{dT_{ap}}{dz} - k_{ea} \frac{dA_a}{dz} \frac{dT_{ap}}{dz} + \pi D_c h_w \left(T_{ap} - T_w \right) - \sum \left(-\Delta H_r \right) \Re_p A_a$

Table 2 below summarizes the one dimensional model formulation and the correlations used in this work for bed hydrodynamics and heat transfer description. Two regions were considered: the central dilute core (“spout”), with upward moving solids entrained by a co current flow of fluid, and the dense phase annular region (“annulus”), with counter current percolation of fluid.

Chemical Scheme

Our main goal was to develop a mathematical model for the simulation of spouted bed reactors operating at high temperature. The process selected in the present work was coal gasification.

The reaction scheme chosen for the modelling consists of the devolatilization process and homogeneous and heterogeneous oxidation and gasification reactions (*Table 3*). The drying process was assumed to occur in parallel with devolatilization, which takes place instantaneously in the gas injection region and produces volatiles (CH_4, H_2, CO_2, CO, H_2O …) and char.

Table 2: Model correlations

	Estimated by
Hydrodynamics	
Region delimitation	
Spout diameter	Wu et al.'s correlation (1987)
Maximum spoutable bed	Wu et al.'s correlation (1987)
Voidage profiles	Correlations of Morgan et al. (1985) and Day (1987)
Stable spouting velocities	
Minimum fluidisation velocity	Littman et al.'s correlation (1981)
Minimum spouting velocity	Wu et al.'s correlation (1987)
Superficial velocities	
Gas in the spout	Continuity balance
Particles in the spout	Morgan et al.'s correlation (1985)
Gas in the annulus	Mamuro and Hattori's correlation (1968)
Particles in the annulus	Continuity balance
Heat transfer	
Annular effective conductivity	Kunii and Smith's correlation (1960)
Gas particle heat transfer	
In the spout	Rowe and Claxton's correlation (1965)
In the annulus	Handley and Heggs's correlation (1968)
Wall heat transfer	Wender and Cooper's correlation (1958)

It was assumed that the total yield of volatiles equalled the volatile content of the coal determined by the proximate analysis. For sake of simplification, it was assumed that the char consists of pure carbon.

The reaction scheme was thus defined by nine reactions (four surface reactions and five gas phase reactions) involving seven chemical species; solid carbon, carbon dioxide, carbon monoxide, methane, water, hydrogen and oxygen.

The chemical system, presented in *Table 3*, has been selected after an exhaustive bibliography analysis on gasification reactions and processes. Since, for each reaction considered, there are different possible kinetic equations in the literature, the criteria of discrimination were the citation frequency, data availability and expression simplicity. In terms of energy balance, the chemical reaction-generated energy was assigned to gas for gas-phase reactions, and to particles for solid surface reactions.

Numerical Solution Scheme

In order to solve the model equations, boundary conditions were specified. Initial oxygen concentration was calculated from the inlet air flow rate. Concerning CO_2, CO, CH_4, H_2O and H_2, it was assumed that the initial concentrations of these species corresponded to volatile products from coal devolatilization and they were estimated from Loison and Chauvin's (1964) empirical correlations.

Thermal boundary conditions were enforced: gas temperatures at the base of the bed (spout and annulus) were assumed equal to inlet gas temperature. Since the particle temperature at z = 0 was unknown, a multi-point boundary value problem had to be solved and it was necessary to specify a temperature condition at the top of the annulus. As Smith et al. (1982) have shown, the condition to be fulfilled is that the top of the annulus and exit spout solids temperatures are equal. The problem was solved using a shooting method associated to an adaptative stiff integration

procedure proposed by Gear (Hindmarsh, 1983)
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Table 3: List of considered homogeneous and heterogeneous reactions.

Reaction	***References for kinetic rate expressions***
Heterogeneous reactions	
(S-1) Char Combustion $C + \Phi O_2 \rightarrow 2(1-\Phi)CO + (2\Phi - 1)CO_2,\ \Phi = 0.8$	Saito et al., 1983
(S-2) Steam gasification $C + \beta H_2O \rightarrow (2-\beta)CO + (\beta - 1)CO_2 + \beta H_2,\ \beta = 1.1$	Matsui et al., 1985
(S-3) Boudouard reaction $C + CO_2 \leftrightarrow 2CO$	Matsui et al., 1987
(S-4) Hydrogen gasification $C + 2H_2 \rightarrow CH_4$	Biba et al., 1978
Homogeneous reactions	
(G-1) Gas combustion $H_2 + 1/2\, O_2 \rightarrow H_2O$	Haslam, 1923
(G-2) Gas combustion $CO + 1/2\, O_2 \rightarrow CO_2$	Tesner, 1960
(G-3) Gas combustion $CH_4 + 1/2\, O_2 \rightarrow CO + 2H_2$	Haslam, 1923
(G-4) Water Gas Shift reaction $CO + H_2O \leftrightarrow CO_2 + H_2$	Biba al., 1978
(G-5) Steam reforming of methane $CH_4 + H_2O \leftrightarrow CO + 3H_2$	Wang et al., 1993

SIMULATION RESULTS AND DISCUSSION

The experimental results obtained by Lucas et al. (1998) and Salam and Bhattacharya (2006) for coal gasification in a spouted bed reactor were adopted to check and validate model predictions. Lucas et al. studied an oxygen-steam gasification experiment for an anthracite coal whereas Salam and Bhattacharya compared air gasification of mangrove charcoal in two different spouted bed configurations. Based on the model described above, the profiles of gas composition, velocities and temperature in the spout and annulus, and finally the overall carbon conversion were calculated and compared to these two experimental conditions *(Table 4)*. The simulation results are in good agreement with the experimental data, except for methane concentration in the case of Salam and Bhattacharya (2006). It seems that our calculated value is too high due to an overestimation of the initial CH4 concentration from Loison et Chauvin's correlation. In Lucas et al.'s case, no visible error arises from this correlation since water is present in large amounts and consumes methane through reaction G-5, which is not the case with Salam and Bhattacharya's data. Different model versions have also been tested according to the heat transfer hypotheses assumed (isothermal or adiabatic reactor, no radiation) and show less accurate results at high temperature.

Table 4: Comparison of predicted and experimental results.

	Lucas et al. (1998) h_c = 0.61 m (RUN A)		***Salam*** and Bhattacharya ***(2006)*** h_c = 0.1m	
Composition at reactor exit (vol. %)	Exp.	Simul.	Exp.	Simul.
CO_2	15.95	17	14.08	12.85
CO	8.72	9.5	13.96	14.1
CH_4	0.18	0.1	0.56	4
H_2O	61.00	60	—	1.36
H_2	7.83	6	12.39	11.6
N_2	6.32	7.4	56.36	56.1
O_2	—	—	2.65	—
Average bed temperature (K)	1198	1190	1252	1265
Carbon conversion (%)	—	62	56.12	56.2

Temperature Profiles

Figure 1 shows the calculated temperature profiles in the spouted bed according to the reference cases; *Figure 1(a)* illustrates data by Lucas et al. (1998) whereas *figure 1(b)* illustrates data by Salam and Bhattacharya (2006).

As shown in *figure 1(a)*, the particle temperature along the spout increases very rapidly in the lower part, reaches a maximum then decreases gradually towards the average bed temperature. This peak results from the coal combustion and the complete consumption of oxygen up to that point. This suggests that the gasification reactions S2 and S3 are much slower than the combustion reaction S1. As long as oxygen is present in the gas phase, there is a significant temperature difference between the coal particles and the gas, since the simultaneous combustion of solid carbon and volatiles generate heat in the solid phase.

Once the oxygen is consumed, the difference between the particle and gas temperatures decreases and becomes negligible, as the gas flowing in the spout is warmed up. In the annulus, the gas and particle temperature profiles are almost identical and equal to the bed average temperature, which can be explained by the excellent mixing and heat transfer that occurs in this region.

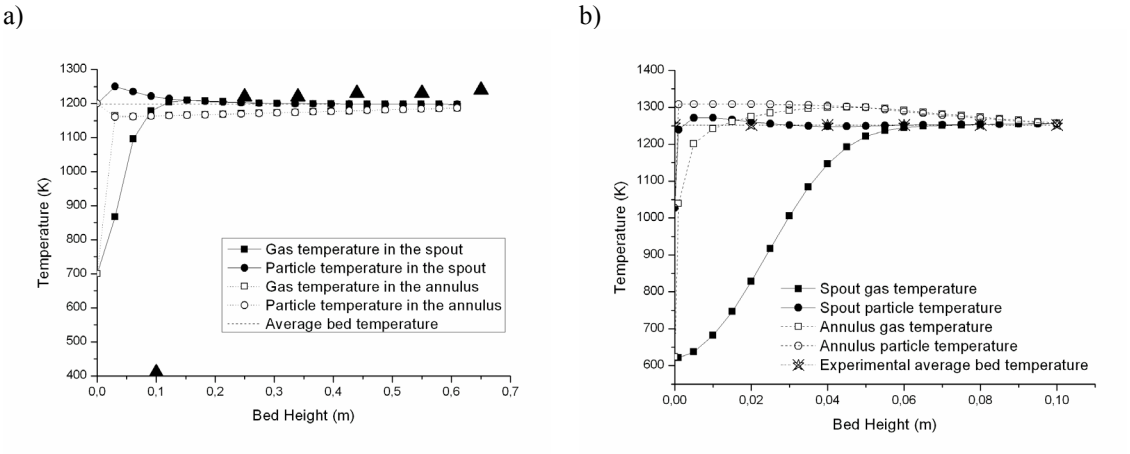

Figure 1: Predicted temperature profiles: a) Lucas et al.'s (1998) data (Run A), b) Salam and Bhattacharya (2006) data.

Similar trends in temperature variations are observed for the data by Salam et al. (*figure 1b*): i) rapid increase of spout particle temperature which corresponds to oxygen combustion, ii) equilibrium between gas and particle temperatures in spout at reactor exit. The particle temperature in the annulus also rises sharply before it slightly decreases with bed height and equals the gas temperature.

Concentration Profiles

The calculated concentration profiles of carbon dioxide, carbon monoxide, hydrogen and steam are illustrated in *figure 2(a)* for the case of Lucas et al. (1998) and in figure 2(b) for the case of Salam and Bhattacharya (2006).

The oxygen profile is not presented since O_2 is quickly consumed at spout entrance and not present in the annulus. The profile evolutions for CO_2, CO, H_2O and H_2 near the inlet results from the competition between the combustion and gasification reactions. As long as oxygen is present, the coal combustion reaction S1 and the volatile combustion reactions (G1 to G3) are preponderant; any methane, hydrogen or carbon monoxide produced by gasification reaction is consumed in the gas phase.

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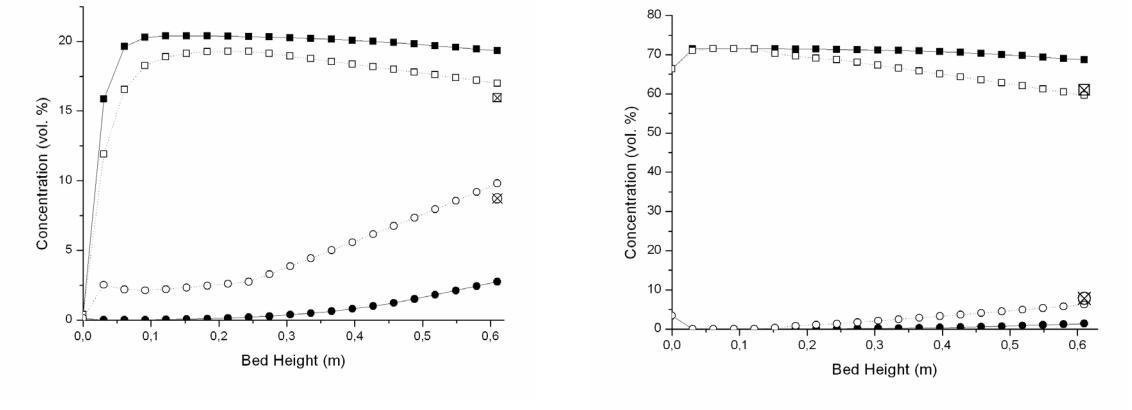
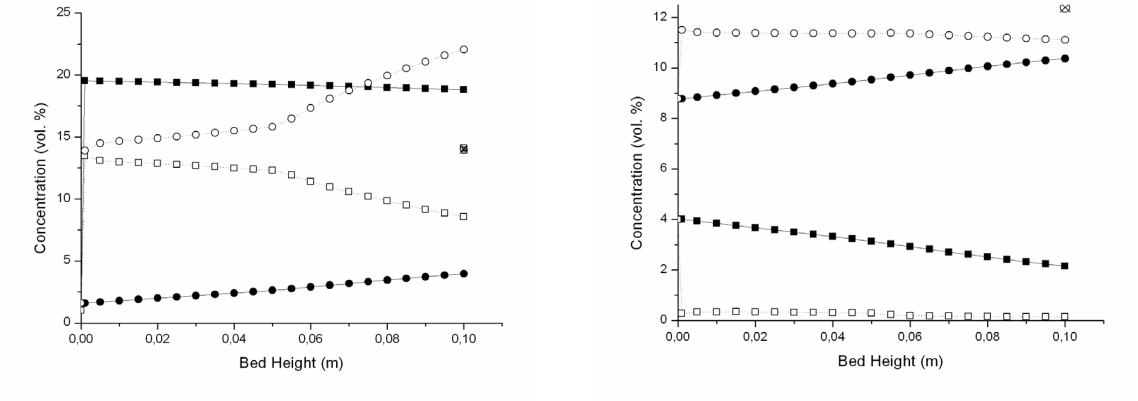
Consequently, in this oxidation zone, the concentrations of carbon dioxide and steam increase, presenting maxima after oxygen depletion. After this point, their profiles decrease slowly as reduction and gasification reactions become more important and carbon monoxide and hydrogen are formed.



a) Lucas et al's (1998) data



b) Salam and Bhattacharya (2006) data

-■- Spout CO_2 concentration
-□- Annulus CO_2 concentration
☒ Average experimental CO_2 concentration
-●- Spout CO concentration
-○- Annulus CO concentration
⊗ Average experimental CO concentration
-■- Spout H_2O concentration
-□- Annulus H_2O concentration
☒ Experimental H_2O concentration
-●- Spout H_2 concentration
-○- Annulus H_2 concentration
⊗ Experimental H_2 concentration

Figure 2: Predicted composition profiles.

Gas species composition variations in the annulus are quite similar to those in the spout. The main difference lies in the fact that carbon monoxide and hydrogen concentrations in the annulus are higher than those in the spout. Conversely, carbon dioxide concentration is slightly lower in the annulus than in the spout.

Such differences are due to the coal gasification reactions that are favoured by higher particle concentration in the annulus. Finally hydrogen profiles show opposite behaviour; hydrogen is produced in the spout and consumed in the annulus. These trends suggest that the spout entry tends to be an oxidizing exothermic region while the annulus behaves as a reducing endothermic region.

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CONCLUSION

In this work, a one dimensional model incorporating hydrodynamics, coupled heat and mass transfer and chemical reactions has been developed and used to simulate coal gasification.

Simulation results have been compared with experimental data of Lucas et al. (1998) and Salam and Bhattacharya (2006). Carbon conversion, bed temperature and exit gas composition predictions have been found in rather good agreement with the experimental data. This model, which takes into account heterogeneous and homogeneous reaction kinetics, has demonstrated its capability to capture all the main chemical and physical processes taking place during coal gasification. Moreover, a rather complete description of heat transfer phenomena seems to be of key importance for an accurate prediction of the main characteristics of a high temperature spouted bed reactor.

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Nomenclature

A	Cross sectional area, m^2
C	Concentration, $mol.m^{-3}$
C_p	Specific heat, $J.m^{-3}.K^{-1}$
D_c	Column diameter, m
d_p	Particle diameter, m
h_{pg}	Gas-particle heat transfer coefficient, $W.m^{-2}.K^{-1}$
h_w	Wall heat transfer coefficient, $W.m^{-2}.K^{-1}$
$-\Delta H_r$	Heat of reaction, $J.mol^{-1}$
k_{ea}	Bed effective conductivity coefficient, $W.m^{-1}.K^{-1}$
r_j	Formation rate of species j ($mol.m^{-3}.s^{-1}$)
$\Re_i$	Global kinetic rate of reaction I ($mol.m^{-3}.s^{-1}$)
T	Temperature, K
U	Gas superficial velocity, $m.s^{-1}$
V	Solid superficial velocity, $m.s^{-1}$
z	Axial coordinate, m

Greek letters and Subscripts

ε	Voidage
ρ	Density, $kg.m^{-3}$
a	Annulus
g	Gas
p	Particle
s	Spout
w	wall